



Dielectric properties of mixed glycine fluoride crystals

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Glycine Hydrogen Potassium Fluoride (GHKF) crystals were grown from aqueous solution by slow evaporation method for the first time. Variation of conductivity with temperature (30°C - 120°C) was measured. The variation in dielectric constant (K) and tangent loss of pressed crystals as a function of frequency in range 400Hz - 1MHz was recorded. The change in dielectric constant with temperature and applied dc bias was studied. Conductivity is found to be very low and decreases drastically with rise in temperature. The dielectric constant is found to be independent of dc bias. Dielectric constant (K) and $\tan \delta$ decreases with frequency to attain fixed value at high frequencies, a typical of a dielectric solid and migrational polarisations. The dielectric constant versus temperature curve exhibits diffused ferroelectric phase transition.

Keywords: Mixed glycine fluoride crystals, dc bias, conductivity, dielectric properties

78 30 -J, 77 22 -d, 72 80 Jc

Glycine Sulphate (TGS) class of ferroelectrics has attracted attention of many [1 - 5] scientists resulting in development of applications of TGS. Also it is well known that by doping such crystals their transition temperature can be controlled. While presence of defects like dislocations and impurities form the cause of their resistivity, changes in dipole moment and domain wall motion govern its dielectric properties.

In new ferroelectric material, glycine hydrogen potassium fluoride (GHKF) crystals were grown in this laboratory for the first time. The physical behaviour of a given material may be characterized by the set of macroscopic measurable quantities such as electrical conductivity, its dielectric constant etc. In fact, these quantities are functions of externally variable parameters such as temperature, pressure, frequency and applied field [6]. The anomaly in these parameters is usually interrelated. The presence of defects influence most of the structural and electrical properties in such solids.

Conduction in solids arises from movement of both the ions and electrons. Proper electric and the ions of foreign inclusions. The dielectric constant at low frequencies depends on factors like ionic, dipole orientation and migrational (space-charge) polarisations. The plot of K against frequency indicates

the contributions of these parameters. The permittivity-frequency relationship describes the dispersion characteristic of a dielectric. A monotonic decrease in permittivity with increase in frequency indicates relaxation dispersion. This is a characteristic of a dipole and migrational polarisation.

The dielectric loss in solid dielectric materials must be considered in connection with their structure. Because solids differ in structure and composition in great varieties of ways, they can display all types of dielectric loss. The dielectric loss further depends on temperature displaying a maximum and a minimum of loss.

Ferroelectric materials are crystalline substances which display a spontaneous polarisation whose direction can be reversed under external influences like an electric field. One more specific parameter of these materials is the ferroelectric Curie temperature. At Curie point, the crystal passes from a ferroelectric to paraelectric phase. The phase transformation is of three types including the diffused one. In the first kind of ferroelectric phase transition, the spontaneous polarisation abruptly drops to zero at the Curie point, the Curie-Weiss temperature being low than the Curie temperature. In the second kind, these two temperatures are equal. In the third (diffused) kind, the definite

transition point is absent and the transition occurs over a broad temperature region (Curie region). In this region, ferroelectric and paraelectric phases are said to coexist.

Hence, studies of interest such as the variation of conductivity with temperature and dielectric constant with temperature, frequency ($\tan \delta$) and applied d.c. bias were carried out on GHKF and are presented here.

Glycine, hydrogen fluoride and potassium fluoride, all AR Grade (Merck, India Ltd) in molar ratio 3 : 0.5 : 0.5 were mixed in distilled water and kept at 35°C and whitish crystals (0.5 - 1.0 cm) were obtained in about 4-5 weeks time. The crystals had many flat faces and lacked good cleavage, hence, they were pressed in form of pellets and subjected to electrical characterization.

Variation of dielectric constant with d.c. bias and frequency and $\tan \delta$ was made using HP 4284 (400Hz- 1MHz) precision LCR-Q meter. For the measurements of conductivity and dielectric constant with temperature, CIE 5125 make digital multimeter with 2000 Megaohms range was employed.

Conductivity (σ) against temperature ($1/T$)

The variation of conductivity with temperature (RT to 120°C) was studied (Figure 1). The overall conductivity of the sample is low as reported by Whipps *et al* [9]. With rise in temperature, it decreases rapidly upto 68°C to become constant. At lower temperatures, impurities are predominant whereas at elevated temperatures, some ions may detach from the sites of crystal lattices [10]. Further, this nature is attributable to a diffused phase transition. The low value of conductivity is associated with the presence of impurities.

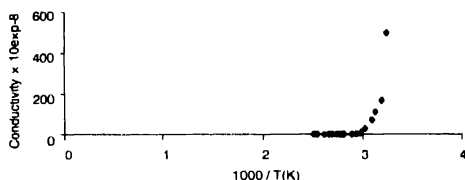


Figure 1. Conductivity versus $1/T$ (K) plot

Dielectric constant Verses d.c. bias (V/d)

Figure 2 shows with application of d.c. bias in range from (0- 63 Volts/cm), the dielectric constant (K) decreases only marginally.

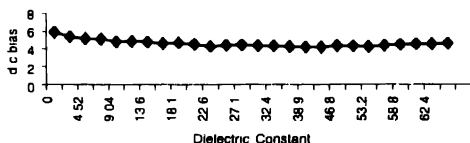


Figure 2. Dielectric constant (K) versus d.c. bias (V/cm)

to become constant, i.e. it is independent of applied d.c. bias. However, the small value of K is ascribed to the domain closure effects seen in such samples [11].

Dielectric constant (K) and $\tan \delta$ with frequency

Pressed specimens contain voids, grain boundaries and defect. The presence of voids decreases the dielectric constant in general [12].

Figures 3 and 4 show the variation of the dielectric constant (K) and tangent loss ($\tan \delta$) factor with frequency (400g- 1MHz). At lower frequencies, K is high and decreases

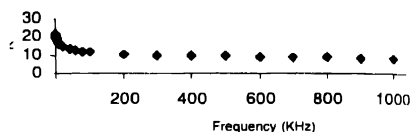


Figure 3. Dielectric constant versus frequency plot

increase in frequency to attain a fixed value at higher frequency. The variation of dielectric constant with frequency is polar dielectric. Moreover, a monotonic decrease in K with increase in frequency (relaxation dispersion) is typical for dipole and migrational polarisation. The value of K is also attributed to defects [13,14]. The nature of Loss tangent curve is a typical dielectric solid. The growth in the loss beyond a minimum is attributed to increase in the leakage loss.

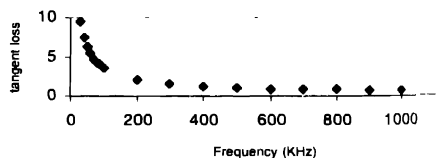


Figure 4. Tangent loss versus frequency plot

Dielectric constant (K) with temperature (°C)

The plot of K against temperature is shown in Figure 5. Dielectric constant increases with temperature to peak at about 40°C thereafter decreases to become constant. The nature of profile is a typical of a diffused phase transition encountered

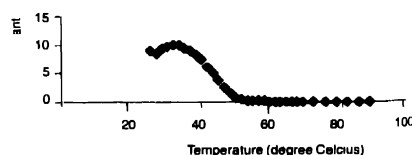


Figure 5. Dielectric constant versus temperature (°C)

piezoelectric materials. Here, the phase transition is said to occur in a broad temperature range with coexistence of the ferroelectric and the paraelectric phases [15].

New mixed glycine fluoride crystals have modified habitats, lack good cleavage planes. The conductivity is low at room temperature and is influenced by presence of impurities and defects. The dielectric constant is found to be independent of applied d.c. field (bias). The variation of dielectric constant with $\tan \delta$ and frequency suggests a dielectric solid with dipole migrational polarisations. The variation of dielectric constant with temperature exhibits diffused phase transition.

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